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Effects of nitrogen and sulphur "stress" treatment on soil acidity and growth response of a Scots pine stand

John Derome[1)], Anna Saarsalmi[2)] and Mikko Kukkola[2)]

[1)] *Rovaniemi Research Unit, Finnish Forest Research Institute, P.O. Box 16, FI-96301 Rovaniemi, Finland (e-mail: John.Derome@metla.fi)*
[2)] *Vantaa Research Unit, Finnish Forest Research Institute, P.O. Box 18, FI-01301 Vantaa, Finland (e-mails: Anna.Saarsalmi@metal.fi, Mikko.Kukkola@metla.fi)*

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In the early 1990s, revitalization fertilization experiments were established in Finland to investigate the possibilities of mitigating the effects of acidifying deposition. Annual treatments (ammonium sulphate) equivalent to 5-fold annual N and S deposition were also included during 1991–2000. The results presented here are from a replicated experiment in a Scots pine stand growing on a relatively infertile, coarse-textured soil in southern Finland. The N + S treatment strongly increased stand growth and litterfall production, indicating that the pines were still suffering from a serious shortage of nitrogen. There were no indications of nitrogen saturation, and no signs of nitrification. The serious leaching losses of Ca and Mg from the surface soil appeared to be primarily due to the addition of large amounts of sulphate. According to the results from the control plots, levels of acidifying N and S deposition in southern Finland had not been high enough to cause a threat to site fertility through soil acidification.

Introduction

During the 1980s and 1990s there was considerable concern in Finland about the possible effects of elevated levels of acidifying nitrogen and sulphur deposition on the health and vitality of the forests. The alarming reports from central Europe about forest dieback caused by acidification and associated nutrient imbalances raised the question of whether Finnish forests were already suffering from the effects of acidifying deposition and, if not, was it likely to become a problem in the near future. In addition to carrying out a number of national and regional projects directed at surveying the health and vitality of the forests and the susceptibility of forest ecosystems to anthropogenic pollution during the late 1980s and early 1990s, the Finnish Forest Research Institute also established a Forest Revitalization Project at the beginning of the 1990s to investigate the long-term effects of slow-release compensation/revitalization fertilizers on forest health and vitality on a number of potentially susceptible sites in Finland (Mälkönen *et al.* 2000). As nitrogen and sulphur deposition in Finland have never reached the high levels experienced in central Europe and southern Scandinavia, the experiments also included a so-called N + S stress treatment. The purpose of this stress treatment was to simulate the long-term effects of increased (ca. 5-fold) nitrogen and sulphur deposition on site fertility and stand growth

in order to assess future trends and to develop means of counteracting the potential acidification threat.

During the past two decades there has been a considerable decrease in acidifying sulphur deposition in Finland, but no corresponding decrease in nitrogen deposition (Kulmala *et al*. 1998, Nordlund 2000). The threat posed by acidifying sulphur deposition is obviously decreasing, but the situation with respect to nitrogen deposition remains unclear. Although the proportion of unfertile, coarse-textured forest soils in Finland is relatively low, less than 10% of the upland forest area, they are frequently located over important groundwater aquifers. These sites are susceptible to the adverse effects of acidifying deposition (Tamminen and Mälkönen 1986), with the associated risk of a deterioration in groundwater quality. Soil acidification, which is normally seen as a decrease in pH, an increase in exchangeable Al concentrations and the loss of exchangeable base cations (primarily Ca and Mg) from the upper soil layers, can occur through a number of mechanisms:

1. An increase in the uptake of nutrient cations (Ca^{2+}, Mg^{2+}, K^{+}, NH_4^{+}) results in the liberation of equivalent amounts of H^{+} ions into the soil solution.
2. The microbial conversion of NH_4^{+} to NO_3, which can take place in forest soil if the NH_4 concentrations are elevated, the pH relatively high (i.e. close to 7), or the C/N ratio below about 25, results in the release of H^{+} ions (Gundersen *et al*. 1998).
3. The leaching of e.g. Ca and Mg in association with inorganic ions such as sulphate or nitrate (Singh *et al*. 1980).

The aims of this study were to investigate the effects of repeated, annual applications of ammonium sulphate, given during a 10-year period, (1) on the chemical properties of the soil and percolation water, and (2) on the nutrient status, growth and litterfall in a Scots pine stand growing on a relatively infertile site in southern Finland. The hypothesis set in the study was that the addition of annual doses of ammonium sulphate, equivalent to approximately 5 times annual deposition of N and S in deposition, would result in soil acidification and subsequently to serious imbalances in the nutrient status of the tree stand and to a deterioration of stand growth on a site that is theoretically extremely sensitive to the detrimental effects of acidifying deposition.

Material and methods

Establishment of the experiments

A field experiment was established in a 45-year-old Scots pine (*Pinus sylvestris*) stand at Hämeenkangas (61°45´N, 22°40´E, 140 m a.s.l.), western Finland, in spring 1991. At the time of establishment the stem number of the stand was 2017 stems ha^{-1}, mean height 9.4 m, stem volume 50 m^3 ha^{-1}, and annual volume increment 3.3 m^3 ha^{-1}. The stand was thinned in 1996 (29% of the total volume) according to the normal guidelines for practical forestry. The stand is growing on a relatively infertile site of the *Calluna vulgaris* forest site type (CT) (Cajander 1949). At the time when the experiment was established the amount of total N in the organic layer was 253 kg ha^{-1} and the C/N ratio was 39 (Mälkönen *et al*. 2000). The effective temperature sum (threshold +5 °C) at the site is 1163 dd., and the long-term mean annual precipitation ca. 600 mm. The ca. 1.5 cm thick humus layer is of the mor type, and the soil type haplic podzol (FAO-UNESCO 1988). The soil texture down to a depth of ca. 50 cm is coarse sorted sand, and below this depth fine sorted sand.

The experiment was laid out in a randomised plot design, with six treatments and three replications of each treatment. The size of the plots was 30 m × 30 m surrounded by a 5-m buffer zone around each plot. The treatments in the experiment consisted of a control (C), a mixture of fast and slow-release mineral fertilizer (MF), MF + nitrogen (MF + N), MF + N + limestone (MF + N + L), a site-specific fertilizer mixture of macro- and micronutrients based on foliar and soil analyses (SSF), and a "stress" treatment consisting of nitrogen and sulphur (N + S). Only the results of the N + S treatment are presented in this paper. The results of the other treatments are presented in Mälkönen *et al*. (2000).

In the N + S treatment, 25 kg N and 30 kg S ha^{-1} were applied annually to the forest floor as ammonium sulphate granules during the 10-year period 1991–2000. In total, 250 kg N ha^{-1} and 300 kg S ha^{-1} were applied. The nitrogen load derived from deposition in western Finland during the same period was ca. 50 kg N ha^{-1} (Nordlund 2000), which means that the total load on the plot during the 10-year period was 300 kg N ha^{-1}. In Finland the ratio between NH_4-N and NO_3-N in deposition is ca. 1:1. The corresponding value for sulphur deposition was ca. 50 kg S ha^{-1} (Nordlund 2000), giving a total sulphur load on the plots of 350 kg S ha^{-1}. The sulphur in deposition in Finland is almost exclusively sulphate.

Tree stand measurement

The tree stand was measured at the time the experiment was established, and again 5 and 10 growing seasons after the start of the experiment. The breast height diameter of all the trees was measured to an accuracy of 1 mm. On each plot at least 30 permanent sample trees representing different size categories were chosen for tree height measurement using a hypsometer to an accuracy of 1 dm. The size categories were determined by first dividing all the trees into five diameter classes, each with the same basal area. Six trees were then randomly selected as sample trees from each size category. The sample trees were used for estimating the height and volume.

In order to study the timing of the response, the additional annual radial growth measurements made 10 years after the start of the experiment were used. The radial growth of five trees on each plot (i.e. 15 per treatment), were chosen randomly to represent five different size categories as described above.

Needle sampling and analysis

Needle samples were collected during the winter before the start of the experiment (1990), and then again in March 1993, October 1995 and November 2000, i.e. 2, 5 and 10 years after the start of the experiment. In 1990 and 1993 needle samples were taken from five and three sample trees, respectively, that were randomly selected from the dominant crown layer on each sample plot. Current needles growing on the third to fifth branch whorl, counting from the top, were sampled on the southern side of the crown. In 1995 and 2000 needle samples were taken from felled sample trees. Current needles were sampled on the middle branch in the upper quarter of the live crown from seven (in 1995) or five (in 2000) sample trees representing different size categories.

The needle samples were dried (40 °C, 48 h) and analysed separately for each tree. Needle unit mass (mg/needle) was also determined separately for each tree. The concentrations of P, K, Ca, Mg, Mn, Cu, Zn, Fe and B were determined on finely ground needles by dry ashing (550 °C for 2 h), followed by extraction with HCl (Halonen *et al*. 1983) and analysis by ICP-AES (TJA, Iris, Advantage). The N concentration was determined on a CHN analyser (Leco). The S concentration was determined on the samples taken before the start of the experiment and on those taken after 10 years only, by wet digestion (HNO_3 + H_2O_2) (Huang and Schulte 1985), followed by analysis by ICP/EAS.

Litterfall sampling and analysis

Tree litterfall was collected using 10 litter traps systematically located on two of the three replicate control and N + S plots. The traps consisted of funnels made of plastic-impregnated, heavy-duty cloth with a collection area of 0.5 m^2 at a height of 1.5 m above ground level. A cotton bag was attached to the bottom of the funnel and changed at each sampling. The litter traps were emptied every second week during April–July, and weekly during August October up until the formation of a permanent snow cover. The traps emptied for the first time in the spring contained litterfall from the preceding winter.

The litter was dried (+40 °C) and separated into a number of fractions: non-green needles, twigs, bark, cones, seeds, flowers and miscellaneous litter and weighed separately. The litter from all the litter traps on each plot was combined to give one composite sample per fraction per

plot per sampling date. When determining the nutrient concentrations of the individual litterfall fractions, samples from two to three different sampling dates were combined. The concentrations of N, P, K, Ca, Mg, Mn, Cu, Zn, Fe and B in the individual litter fractions were determined as described above for living needles.

Soil sampling and analysis

Soil samples were taken from the organic layer (F + H) and the mineral soil at depths of 0–5 and 5–10 cm in the year 2000. The samples were taken systematically at 25 sampling points on each plot, and the samples from the same plot were bulked by layer. The organic layer samples were taken using a cylinder (d = 58 mm) and the mineral soil samples using an auger (d = 21 mm). The thickness of the organic layer was measured in conjunction with sampling. The soil samples were dried in a ventilated chamber at a temperature of 30–40 °C. The organic layer samples were ground in a mill with a 2-mm bottom sieve, and the mineral soil samples were passed through a 2-mm sieve to remove stones and larger roots. Soil pH was determined in a water suspension with a ratio of 15 ml of sample and 25 ml of distilled water. Total N and C were determined on the organic layer and mineral soil samples on a CHN analyser (Leco). Exchangeable base cations (Ca, Mg, K, Na) were determined by extraction with 0.1 M $BaCl_2$ using a ratio of 15 ml of sample and 150 ml of extractant. The suspensions were left to stand overnight before being shaken for 1 h and then filtered. The concentrations of Ca, Mg, K, Na and Al were determined by ICP-AES (ARL 3580). Exchangeable acidity (EA) was determined by titrating the $BaCl_2$ extract with 0.05 M NaOH to pH 7.0.

Soil solution sampling and analysis

Percolation water was collected at 4-week intervals during the snowfree period (on average May–October) during 1991–1999 using zero-tension lysimeters installed at a depth of 20 cm below the ground surface. The construction and installation of the lysimeters are described in detail in Derome *et al*. (1991). 20 lysimeters/plot were installed on two of the three replicate control plots and on two of the three replicate N + S plots. The samples from the individual lysimeters were bulked to give one sample per plot per sampling date. On arrival at the laboratory, the pH of the samples was measured and the samples then filtered through a 0.45 μm membrane (Schleicher & Schull) filter. Nitrate and SO_4 were determined by ion chromatography (Dionex Corporation), and Ca, Mg, K and Al by ICP-AES.

Calculation of the results and statistical analyses

The statistical significance of the differences in needle nutrient concentrations, litterfall concentrations, soil solution chemistry and in soil chemical properties between the treatments was tested using the independent-samples t-test. The equality of variances was tested with Levene's test. Pooled or separate variances were used depending on the equality of the variances.

The stand characteristics at each measurement round were calculated using the KPL calculation programme for sample plots (Heinonen 1994). A function based on breast height diameter and height was used for calculating the volume of the sample trees (Laasasenaho 1982). Plotwise height and volume equations based on dbh were then calculated using the data from the sample trees in order to estimate the height and volume of the other trees. The growth during both 5-year periods was calculated as the difference between consecutive measurements (the same trees at the start and at the end of each period; also trees which had died during each period (0%–6%) were included). Statistical significance of the differences in volume growth between the treatments was tested using analysis of variance instead of the t-test because the initial stand volume was used as a covariate to adjust for differences in pre-treatment growth. Bonferroni's test was used to test the equality of the treatment means.

The mass of the litterfall fractions (non-green needles and other litter) per unit area was calculated by dividing the mass of the collected litter-

fall fraction by the surface area of the litter traps. The amounts of nutrients reaching the ground as needle litterfall were calculated by multiplying the nutrient concentrations of the needle litter for each sample period by the corresponding mass of the needle litter. The amounts of nutrients reaching the ground as other litter were calculated by multiplying the nutrient concentrations of the litter fraction in question for each sample period by the corresponding mass of litter and summing up the amounts of individual litter fractions. In this paper, we present the results separately for non-green needles (later called needle litterfall) and all the other fractions (other litterfall) combined.

Annual mean values (and standard errors of the mean) were calculated for the pH and Ca, Mg, Al, SO_4-N and NO_3-N concentrations in percolation water for the control and N + S treatments separately. As there was considerable year-to-year variation in these parameters, the effect of the N + S treatment is presented in the figures as the difference compared to the control treatment. Statistically significant differences between the treatments (N + S and control) were tested using the *t*-test. Significant differences are marked on the relevant figures using an asterisk ($p < 0.05$).

Results and discussion

In this experiment, N and S were applied to the forest floor in the form of ammonium sulphate at doses approximately 5-fold the input of N and S into the forests from deposition. It is clear, however, that N and S applied directly to the soil is not the same as N and S deposition falling onto the tree canopies and, indirectly, onto the forest floor. There are interactions and chemical transformations between the N compounds in deposition and the tree foliage (N uptake by the needles) and the lichens and microbial communities present in the foliage. Under Finnish conditions, a varying proportion of the N, for instance, is retained in the canopy (Lindroos *et al*. 2007). From the point of view of the effect of N and S on soil processes, the amounts of N and S applied in this study are therefore even more than 5-fold the input from deposition. However, it is clear that the amounts of N and S applied in this experiment are still well below the inputs recorded in southern Scandinavia and central Europe.

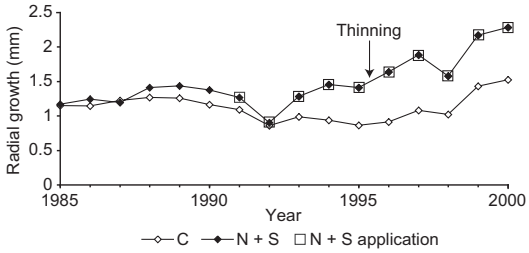

Fig. 1. Radial growth response of Scots pine to the C and N + S treatments. C = control treatment, N + S = annual application of ammonium sulphate.

Growth response of the trees

The annually applied N + S treatment had an increasing effect on radial growth, indicating that N is still the growth-limiting factor on this site (Fig. 1). However, it is not possible on the basis of our material to distinguish between the positive effect of N and the potentially negative, indirect effect of S, because the experiment did not include separate N and S treatments. The effect of small annual nitrogen additions on tree growth started slowly compared to the effects of normal forest fertilization with much larger doses. After the initial 4–5-year period, the cumulative effect of repeated doses reached a relatively constant level (Fig. 1). On the control plots, the annual volume growth was 3.7 and 3.0 m^3 ha^{-1} yr^{-1} during the first and second 5-year period, respectively (Table 1). The absolute increase in volume growth caused by the addition of N + S was 1.7 and 2 m^3 ha^{-1} yr^{-1} during the first and second 5-year period, respectively. This is relatively low compared to the growth increases obtained in pine stands recommended for production fertilization (Kukkola and Saramäki 1983, Saarsalmi and Mälkönen 2001). This is, however, expected because of the low initial volume growth level. On the other hand, the relative growth increase was large, 44% and 67% in the N + S treatment as compared with that in the control plots during the first and second 5-year periods, respectively.

The study site is relatively infertile and located on sorted, coarse-textured soil. Fertilization on such sites is not normally recommended owing to the low wood yield and the potential problem of the fertilizers leaching into the groundwater (Paavilainen 1979). However, the results obtained in this study are interesting in the light of these recommendations because the growth reaction was reasonable and there were no significant signs of nitrate leaching. Furthermore, the nitrogen fertilizers nowadays used in practical forest fertilization do not contain sulphate.

Nutrient status of the trees

At the beginning of the study the needle N concentrations were low (Table 2) and typical for pine stands growing on dry, nutrient poor sites (Jukka 1988). Compared with the mean concentrations for 98 pine stands (years 1987–1989) in different parts of Finland (Raitio *et al*. 2000), the needle B and Mn concentrations were higher at the beginning of the experiment, the N, Ca, S, Fe and Zn concentrations were relatively similar, and the concentrations of K, Mg, P and Cu were lower.

There were significant differences between the needle nutrient concentrations in different years on the control plots. In 1993 the needle P concentrations, and in 1995 both the needle N and P concentrations, were even below the deficiency level of 10.9 and 1.3 g kg^{-1}, respectively, for pine stands growing on this site type (Jukka 1988). In 2000, however, the needle N and P concentrations on the control plots had significantly increased. This is partially due to the heavy thinning carried out in 1996. Significant year-to-year variation in needle nutrient concentrations have also been reported by Helmisaari (1990) and Raitio *et al*. (2000).

The N + S treatment resulted in increased N concentrations in the needles, the response apparently starting already after 2 years (Table 2). Despite this response, the needle N concentrations after 5 years were low and even below the nitrogen deficiency level. The annual N + S treatment had a decreasing effect on the foliar Mg, B, Mn and Zn concentrations after 5 and 10 years. The needle S concentrations were not analysed until 10 years after the start of the

Table 1. Annual stand volume growth (m^3 ha^{-1}, range in parentheses) after the first (1990–1995) and second 5-year period (1996–2000). C = control treatment, N + S = annual application of ammonium sulphate. One-way ANOVA on annual stand volume growth with the stand volume at establishment of the experiment (in 1990) as covariate.

Period		C			N + S	
First 5-year period		3.68 (3.38–3.98)			5.30 (5.00–5.60)	
Second 5-year period		3.04 (2.61–3.47)			5.08 (4.65–5.51)	
Period	Source	Type III sum of squares	df	Mean square	*F*	*p*
First 5-year period	Corrected model	12.4[a)]	2	6.18	248	0.000
	Intercept	0.20	1	0.20	8.0	0.067
	Volume 1990	4.70	1	4.70	188	0.001
	Treatment	3.51	1	3.51	141	0.001
	Error	0.08	3	0.02		
	Total	133	6			
	Corrected total	12.4	5			
Second 5-year period	Corrected model	10.9[b)]	2	5.45	99.0	0.002
	Intercept	0.76	1	0.76	13.8	0.034
	Volume 1990	1.91	1	1.91	34.6	0.010
	Treatment	3.12	1	3.12	56.7	0.005
	Error	0.17	3	0.06		
	Total	110	6			
	Corrected total	11.1	5			

[a)] R^2 = 0.994 (adjusted R^2 = 0.990). [b)] R^2 = 0.985 (adjusted R^2 = 0.975).

experiment. At that time there was a significant increase in needle S concentrations on the N + S treated plots.

Litterfall

On an annual basis, needle litterfall accounts for the main part (ca. 70%) of the total litterfall in Scots pine stands (Mälkönen 1974, Flower-Ellis 1985, Finér 1996, Starr *et al*. 2005). Environmental factors such as site fertility (Albrektson 1988, Vose and Allen 1991) and weather conditions, especially the air temperature (Flower-Ellis 1985, Kouki and Hokkanen 1992, Finér 1996, Saarsalmi *et al*. 2007), cause considerable between-year variation in litterfall production. In our study, the annual needle litterfall production varied on the control plots between 62 to 104 g m^{-2} and accounted for on average of 71% of the total litterfall production (Fig. 2). A temporary drop in litterfall production in 1996 was followed by a rapid increase in litterfall production in the years following thinning.

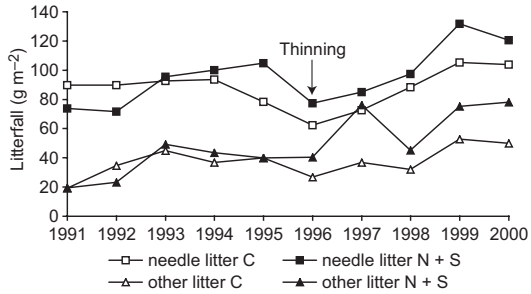

Fig. 2. Annual litterfall of the Scots pine stand during 1991–2000. C = control treatment, N + S = annual application of ammonium sulphate.

Changes in nutrient availability associated with fertilization along natural fertility gradients can increase needle litterfall production (Miller *et al*. 1976, Lim and Cousens 1986, Vose and Allen 1991). However, according to a number of studies, fertilization initially results in relatively

Table 2. Total nutrient concentrations and needle mass (SE in parentheses) in current needles before the start of the experiment (1990), and two (1993), five (1995) and ten (2000) years after the start of the experiment. Values in boldface differ significantly ($p < 0.05$) from the control. C = control treatment, N + S = annual application of ammonium sulphate. $n = 3$.

	Treatment	1990	1993	1995	2000
N (g kg^{-1})	C	11.7 (0.1)	11.6 (0.2)	9.6 (0.3)	13.1 (0.5)
	N + S	11.8 (0.5)	**13.2** (0.5)	10.4 (0.2)	14.4 (0.4)
P (g kg^{-1})	C	1.40 (0.02)	1.14 (0.06)	1.16 (0.02)	1.49 (0.05)
	N + S	1.49 (0.03)	1.16 (0.05)	1.23 (0.07)	1.47 (0.03)
K (g kg^{-1})	C	4.90 (0.42)	3.71 (0.20)	4.15 (0.11)	5.33 (0.11)
	N + S	4.47 (0.15)	3.45 (0.06)	4.41 (0.20)	5.40 (0.09)
Ca (g kg^{-1})	C	1.97 (0.07)	1.21 (0.08)	1.87 (0.13)	2.20 (0.23)
	N + S	1.91 (0.16)	1.42 (0.04)	1.62 (0.15)	1.91 (0.19)
Mg (g kg^{-1})	C	0.98 (0.06)	0.74 (0.05)	1.07 (0.06)	1.12 (0.04)
	N + S	0.77 (0.05)	0.73 (0.05)	0.84 (0.08)	0.96 (0.08)
S (g kg^{-1})	C	1.07 (0.04)			0.91 (0.04)
	N + S	1.18 (0.05)			**1.10** (0.03)
B (mg kg^{-1})	C	16.3 (3.3)	13.8 (1.8)	16.2 (0.6)	16.8 (1.1)
	N + S	14.1 (0.4)	12.8 (1.3)	**11.3** (1.1)	**8.3** (1.8)
Cu (mg kg^{-1})	C	2.3 (0.3)	6.0 (0.3)	4.0 (0.3)	3.4 (0.1)
	N + S	2.2 (0.5)	6.0 (0.3)	4.1 (0.1)	3.7 (0.1)
Fe (mg kg^{-1})	C	47 (2)	41 (4)	37 (3)	43 (1)
	N + S	48 (2)	36 (1)	37 (1)	45 (2)
Mn (mg kg^{-1})	C	529 (68)	211 (2)	302 (18)	373 (43)
	N + S	589 (30)	244 (5)	239 (24)	285 (16)
Zn (mg kg^{-1})	C	41 (2)	40 (4)	44 (4)	64 (4)
	N + S	42 (1)	36 (2)	34 (2)	58 (4)
Needle mass (mg $needle^{-1}$)	C	12.1 (0.9)	10.2 (0.2)	13.1 (0.4)	15.7 (0.9)
	N + S	10.6 (0.4)	10.1 (1.6)	**16.1** (0.9)	17.3 (0.8)

rapid decreases in needle litterfall (Miller *et al*. 1976, Miller and Miller 1976, Lim and Cousens 1986, Mälkönen *et al*. 2000). This decrease in litterfall production is explained by increased longevity of the older needles (e.g. Miller *et al*. 1976). The response of the living needle biomass to fertilization can be seen as an increase in needle litterfall usually two to four years after fertilization (Miller *et al*. 1976, Albrektson *et al*. 1977, Mälkönen *et al*. 2000). In our study, litterfall was lower on the N + S plots during the two first years after the start of the treatment (Fig. 2). However, the treatment subsequently increased litterfall after the fourth year onwards. In 2000, the annual needle litterfall was 15% higher on the N + S plots than on the control plots.

In this study, the response to fertilization of the nutrient concentrations in needle litterfall was closely connected to the response in the living needles. The N + S treatment had an increasing effect on needle N concentrations but a decreasing effect on Mg, B, Mn and Zn concentrations in the needle litter (Table 3). Also in other studies, the N concentration of pine needle litter has been reported to increase after N application (Birk and Vitousek 1986, Finér 1996).

Nitrogen and Ca were the major nutrients in needle litterfall. A total of 6.9 and 8.4 g N m^{-2}, and 3.6 and 4.1 g Ca m^{-2} were returned to the forest floor in total litterfall during the course of the study on the control and N + S treated plots, respectively. During the study, a total of 0.40 and 0.42 g Mg m^{-2} was returned to the forest floor on the control and N + S treated plots, respectively.

Acidity and nutrient status of the soil

The exchangeable K and Mg concentrations (Table 4) in the organic layer were approximately one half the mean values for CT sites in southern Finland, and the exchangeable Ca concentrations correspondingly 60% (Tamminen 1991). These very low concentrations are primarily due to the historical occurrence of forest fires in the area, and subsequent losses of e.g. Mg from the topsoil (Raitio 1991). Base saturation values were extremely low, and well below the range (50%–95%) for the organic layer and close to the lower limit for mineral soil (ca. 5%) reported by Tamminen and Starr (1990) for forested mineral soils in southern Finland. This site is therefore likely to be extremely susceptible to the detrimental effects of acidifying nitrogen and sulphur deposition (Tamminen and Mälkönen 1986).

The annually applied N + S treatment caused a significant decrease in the exchangeable Ca and Mg concentrations in the organic layer, as well as in the exchangeable Mg and K concentrations in the 0–5 cm mineral soil layer (Table 4). A decrease in Ca, Mg and K concentrations in the surface layers of forest soils can be caused either by increased uptake by the tree stand, or by increased leaching losses from the surface soil, e.g. as a result of increased acidification. An increase in the uptake of Ca, Mg and K by the tree stand does not seem to be the main cause of the loss of Ca and K, and especially of Mg, from the surface soil layers: in fact there was a significant drop in the foliar Mg concentrations but no change in the foliar Ca or K concentrations. However, owing to the very strong positive response (44%–67%) of stand growth to the treatment, and increase in needle mass, we can assume that the uptake of Ca, Mg and K has increased to some extent.

Judging by the strong increase in stand growth and increase in foliar N concentrations, a considerable proportion of the NH_4^+ cations given in the treatment has been taken up by the stand. The microbial conversion of NH_4^+ to NO_3, which can take place in forest soil if the NH_4 concentrations are elevated, the pH relatively high (i.e. close to 7), or the C/N ratio below about 25, results in the release of H^+ ions (Gundersen *et al*. 1998). In this study, however, the 5–10 cm mineral soil layer was the only layer where the pH and C/N ratio values came close to fulfilling the requirements for nitrification. However, there were no statistically significant indications of increased nitrate concentrations at a depth of 20 cm on the N + S plots (*see* next section). One of the main processes contributing to soil acidification in soils subjected to elevated levels of nitrogen and sulphur compounds is the leaching of e.g. Ca and Mg in association with sulphate ions (Singh *et al*. 1980). Forest soils contain relatively low concentrations of inorganic anions (e.g. SO_4^{2-}, NO_3^-, PO_4^{3-}), and the leaching of cations from the surface layers of

Table 3. Mean annual nutrient concentrations (SE in parentheses) in needle litter (weighted with litterfall production) during 1991–2000. Values in boldface differ significantly ($p < 0.05$) from the control. C = control treatment, N + S = annual application of ammonium sulphate. $n = 2$.

Year	Treatment	N (g kg^{-1})	P (g kg^{-1})	K (g kg^{-1})	Ca (g kg^{-1})	Mg (g kg^{-1})	B (mg kg^{-1})	Cu (mg kg^{-1})	Fe (mg kg^{-1})	Mn (mg kg^{-1})	Zn (mg kg^{-1})
1991	C	5.02 (0.12)	0.35 (0.00)	0.70 (0.01)	3.61 (0.34)	0.30 (0.02)	9.76 (0.74)	3.43 (0.20)	102 (2)	565 (56)	123 (3)
	N + S	5.58 (0.13)	0.34 (0.00)	0.70 (0.01)	3.80 (0.37)	0.29 (0.02)	7.64 (0.80)	3.36 (0.21)	95 (2)	565 (60)	126 (3)
1992	C	5.30 (0.09)	0.38 (0.01)	0.73 (0.01)	3.13 (0.34)	0.32 (0.01)	7.38 (0.23)	4.04 (0.43)	144 (6)	434 (40)	174 (14)
	N + S	5.11 (0.12)	0.37 (0.01)	0.76 (0.01)	3.81 (0.43)	0.32 (0.02)	6.90 (0.28)	4.37 (0.55)	135 (8)	477 (51)	164 (17)
1993	C	6.02 (0.10)	0.61 (0.03)	0.86 (0.02)	3.31 (0.30)	0.37 (0.02)	8.01 (0.29)	7.24 (0.08)	160 (3)	485 (52)	61 (5)
	N + S	6.42 (0.09)	0.59 (0.03)	0.85 (0.02)	3.71 (0.30)	0.36 (0.02)	6.85 (0.29)	**5.80** (0.08)	**139** (3)	498 (52)	57 (4)
1994	C	4.85 (0.03)	0.48 (0.02)	0.85 (0.02)	3.20 (0.29)	0.35 (0.02)	7.51 (0.67)	4.12 (0.23)	107 (5)	494 (36)	58 (0)
	N + S	5.11 (0.03)	0.42 (0.02)	0.78 (0.02)	3.42 (0.28)	0.33 (0.02)	5.50 (0.65)	3.52 (0.22)	103 (5)	476 (35)	**49** (0)
1995	C	5.37 (0.06)	0.54 (0.02)	0.97 (0.01)	2.76 (0.22)	0.32 (0.02)	7.40 (0.42)	4.05 (0.17)	106 (0)	401 (44)	54 (2)
	N + S	5.47 (0.05)	0.47 (0.02)	0.92 (0.01)	2.93 (0.19)	0.30 (0.01)	5.91 (0.37)	4.38 (0.15)	**87** (0)	378 (38)	**44** (1)
1996	C	5.04 (0.07)	0.41 (0.01)	0.83 (0.01)	3.04 (0.22)	0.33 (0.02)	7.74 (0.34)	3.29 (0.21)	73 (3)	451 (56)	55 (2)
	N + S	**5.46** (0.06)	0.38 (0.01)	0.79 (0.01)	2.93 (0.20)	0.28 (0.02)	**5.36** (0.31)	3.62 (0.19)	65 (3)	380 (50)	**42** (2)
1997	C	4.95 (0.11)	0.52 (0.03)	0.99 (0.01)	3.17 (0.22)	0.40 (0.02)	8.86 (0.33)	5.07 (0.36)	99 (5)	475 (59)	55 (2)
	N + S	5.45 (0.10)	0.45 (0.03)	0.96 (0.01)	3.01 (0.20)	0.33 (0.02)	**6.22** (0.30)	5.04 (0.33)	85 (5)	386 (55)	**43** (2)
1998	C	5.92 (0.12)	0.58 (0.01)	0.80 (0.02)	2.99 (0.24)	0.38 (0.02)	8.06 (0.25)	3.94 (0.19)	98 (8)	439 (61)	54 (2)
	N + S	6.45 (0.11)	0.53 (0.01)	0.79 (0.02)	2.90 (0.23)	0.33 (0.02)	**6.01** (0.24)	3.65 (0.18)	76 (8)	348 (58)	**43** (2)
1999	C	4.81 (0.10)	0.56 (0.01)	0.90 (0.03)	3.34 (0.22)	0.36 (0.01)	7.84 (0.27)	2.80 (0.09)	96 (1)	491 (63)	62 (1)
	N + S	5.35 (0.09)	**0.47** (0.01)	0.88 (0.03)	3.02 (0.20)	**0.31** (0.01)	**5.26** (0.24)	2.81 (0.08)	**84** (1)	357 (56)	**47** (1)
2000	C	5.04 (0.17)	0.51 (0.03)	0.72 (0.01)	3.18 (0.14)	0.31 (0.02)	7.55 (0.43)	2.71 (0.40)	91 (5)	463 (53)	62 (4)
	N + S	5.81 (0.16)	0.45 (0.03)	0.72 (0.01)	2.76 (0.13)	0.27 (0.02)	5.41 (0.40)	2.74 (0.37)	81 (4)	320 (49)	42 (84)

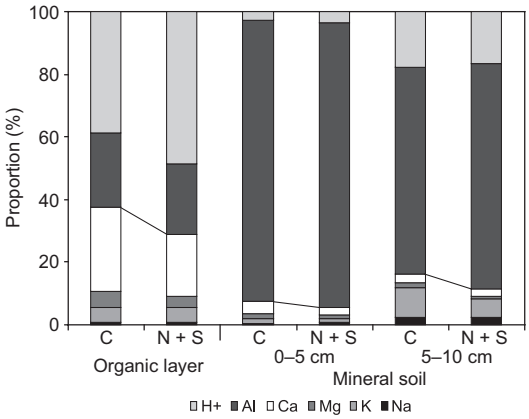

Fig. 3. Effect of the annually applied ammonium sulphate (N + S) treatment on the proportion of base cations (Ca^{2+}, Mg^{2+}, K^{+}, Na^{+}) and acidic cations (H^{+}, Al^{3+}) on cation exchange sites in the organic and 0–5 and 5–10 cm mineral soil layers 10 years after application of the treatment. C = control treatment. The area below the line joining the pairs of columns corresponds to the base saturation value (BS as %).

podzolic forest soils is restricted by the fact that the negatively charged organic matter molecules in the solid phase of the soil are the predominant anions. The annual addition of relatively large amounts of sulphate in the N + S treatment is most probably the primary reason for the loss of Ca, Mg, K and even Al from the surface soil layers (Table 4).

The BS values, which were already extremely low in both the organic layer and the uppermost mineral soil layers on this site, have been strongly decreased by the N + S treatment (Fig. 3). The main reason for the decrease in BS was the loss of exchangeable Ca and Mg and their replacement by H^{+} in the organic layer, and corresponding replacement by Al^{3+} in the 5–10 cm mineral soil layer. It is clear that the N + S treatment has further reduced the capacity of the soil to counteract normal soil acidification resulting e.g. from stand growth.

Percolation water quality

The N + S treatment brought about a consistent significant decrease in pH of about 0.3 pH units in percolation water at a depth of 20 cm two years after the treatment was first applied (Fig. 4a and Table 5). This was in good agreement with the decrease in pH found in the 5–10 cm mineral soil layer. There was a relatively strong increase in the Ca concentrations (Fig. 4b) in a number of years, a strong increase in the SO_4-S concentrations (Fig. 4c), a slight increase in the

Table 4. Nutrient status (total carbon (C) and nitrogen (N), C/N ratio, exchangeable Ca, Mg and K, cation exchange capacity (CEC)) and acidity parameters (pH, exchangeable Al, exchangeable acidity (EA), base saturation (BS)) (± SE) in the organic and uppermost mineral soil layers in the control (C) and N + S treatment 10 years after the experiment was established. Statistical significance: * $p = 0.05$, ** $p = 0.01$, and *** $p = 0.001$, $n = 3$.

Parameter	Organic layer			Mineral soil			
	Typical mean and range[1]	C	N + S	0–5 cm		5–10 cm	
				C	N + S	C	N + S
pH (H_2O)	3.69 (3.4–4.5)	3.41 ± 0.02	3.38 ± 0.01	4.15 ± 0.14	3.90 ± 0.04	4.81 ± 0.02	4.50 ± 0.02***
Total C (%)		42.5 ± 1.5	45.0 ± 1.9	2.37 ± 0.21	1.92 ± 0.24	1.70 ± 0.16	1.53 ± 0.04
Total N (g kg^{-1})		12.0 ± 0.3	13.1 ± 0.4	0.74 ± 0.03	0.70 ± 0.06	0.64 ± 0.04	0.64 ± 0.03
C/N ratio		35.5 ± 0.6	34.4 ± 0.3	30.7 ± 1.4	27.1 ± 1.1	27.1 ± 2.2	24.0 ± 0.8
Ca (mg kg^{-1})	1800 (500–3100)	1114 ± 105	723 ± 15*	24.7 ± 5.1	14.4 ± 4.4	6.06 ± 0.95	5.91 ± 1.13
Mg (mg kg^{-1})	250 (70–450)	133 ± 10	82 ± 4**	5.02 ± 0.21	3.67 ± 0.36*	2.54 ± 0.17	2.13 ± 0.18
K (mg kg^{-1})	760 (180–1200)	387 ± 29	333 ± 8	18.3 ± 1.3	13.4 ± 1.1*	11.6 ± 0.9	7.6 ± 1.33
Al (mg kg^{-1})		441 ± 19	376 ± 17	246 ± 12	237 ± 30	92 ± 17	114 ± 6
EA (meq kg^{-1})		130 ± 2	131 ± 0	28.2 ± 1.3	27.4 ± 1.4	11.2 ± 1.8	13.6 ± 0.5
CEC (meq kg^{-1})		208 ± 5	184 ± 1*	30.4 ± 1.4	29.0 ± 1.5	12.1 ± 1.9	14.4 ± 0.5
BS (%)		37.4 ± 2.3	28.7 ± 0.5*	7.45 ± 0.65	5.47 ± 0.90	7.78 ± 0.50	5.60 ± 0.58*

[1] Tamminen 1991.

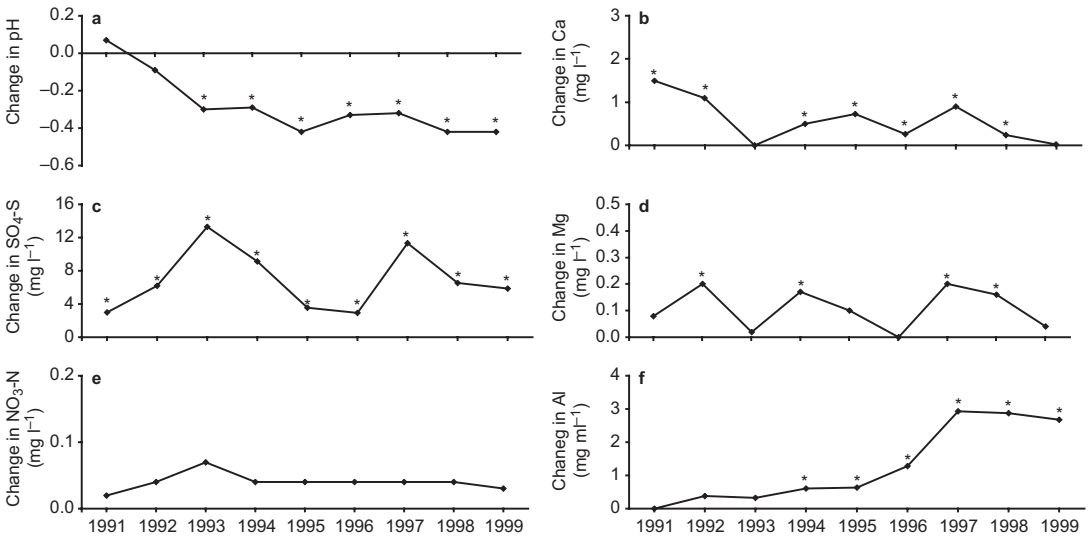

Fig. 4. Change in (**a**) pH and the concentrations of (**b**) Ca, (**c**) SO_4-S, (**d**) Mg, (**e**) NO_3-N and (**f**) Al in percolation water collected at a depth of 20 cm on the plots given the annual N + S treatment compared to the situation on the control plots during 1991–1999. * = significant at $p < 0.05$.

Table 5. Annual mean pH and Ca, Mg, NO_3-N, SO_4-S and Al concentration (± SE) in percolation water collected at a depth of 20 cm during 1991–1999. C = control treatment, N + S = annual application of ammonium sulphate. $n = 2$.

Year	pH		Ca (mg l^{-1})		Mg (mg l^{-1})		K (mg l^{-1})	
	C	N + S	C	N + S	C	N + S	C	N + S
1991	4.66 ± 0.03	4.73 ± 0.04	1.18 ± 0.23	2.67 ± 0.28	0.34 ± 0.03	0.57 ± 0.07	1.43 ± 0.14	1.72 ± 0.22
1992	4.63 ± 0.03	4.54 ± 0.05	0.61 ± 0.23	1.70 ± 0.18	0.19 ± 0.03	0.39 ± 0.04	1.22 ± 0.14	1.32 ± 0.18
1993	4.81 ± 0.06	4.51 ± 0.09	0.30 ± 0.04	0.30 ± 0.06	0.18 ± 0.02	0.20 ± 0.03	1.31 ± 0.08	1.45 ± 0.12
1994	4.81 ± 0.11	4.52 ± 0.13	1.50 ± 0.22	2.00 ± 0.15	0.20 ± 0.03	0.37 ± 0.08	1.27 ± 0.10	1.41 ± 0.11
1995	4.98 ± 0.07	4.56 ± 0.17	0.86 ± 0.07	1.53 ± 0.19	0.23 ± 0.02	0.33 ± 0.04	1.44 ± 0.17	1.53 ± 0.15
1996	4.71 ± 0.10	4.38 ± 0.03	0.57 ± 0.05	0.83 ± 0.09	0.24 ± 0.05	0.24 ± 0.03	1.18 ± 0.10	0.80 ± 0.12
1997	4.69 ± 0.06	4.37 ± 0.05	0.42 ± 0.06	1.32 ± 0.29	0.16 ± 0.02	0.36 ± 0.09	0.59 ± 0.06	0.28 ± 0.36
1998	4.67 ± 0.08	4.25 ± 0.04	0.26 ± 0.04	0.50 ± 0.08	0.10 ± 0.01	0.26 ± 0.07	0.40 ± 0.07	0.30 ± 0.05
1999	4.65 ± 0.05	4.23 ± 0.03	0.33 ± 0.04	0.35 ± 0.04	0.12 ± 0.02	0.16 ± 0.02	2.02 ± 1.32	0.47 ± 0.12

	NO_3-N (mg l^{-1})		SO_4-S (mg l^{-1})		Al (mg l^{-1})	
	C	N + S	C	N + S	C	N + S
1991	0.01 ± 0.01	0.03 ± 0.01	4.13 ± 0.42	9.77 ± 1.96	0.37 ± 0.05	0.27 ± 0.04
1992	0.01 ± 0.00	0.05 ± 0.02	2.78 ± 0.40	6.98 ± 1.07	0.32 ± 0.05	0.70 ± 0.11
1993	0.01 ± 0.01	0.08 ± 0.03	1.70 ± 0.12	15.0 ± 2.93	0.43 ± 0.04	0.75 ± 0.13
1994	0.02 ± 0.01	0.06 ± 0.02	1.90 ± 0.11	11.0 ± 0.75	0.28 ± 0.03	0.89 ± 0.18
1995	0.01 ± 0.01	0.05 ± 0.02	2.01 ± 0.09	6.18 ± 0.57	0.18 ± 0.03	0.88 ± 0.28
1996	0.02 ± 0.01	0.06 ± 0.02	1.80 ± 0.10	4.73 ± 0.25	0.26 ± 0.03	1.54 ± 0.13
1997	0.01 ± 0.00	0.05 ± 0.02	1.84 ± 0.11	13.2 ± 3.89	0.41 ± 0.03	3.34 ± 0.69
1998	0.01 ± 0.00	0.05 ± 0.02	1.79 ± 0.15	8.31 ± 0.62	0.34 ± 0.04	3.21 ± 0.30
1999	0.01 ± 0.00	0.04 ± 0.02	2.37 ± 0.30	8.25 ± 0.60	0.51 ± 0.06	3.19 ± 0.28

Mg concentrations (Fig. 4d), a very small but consistent increase in the NO_3-N concentrations (Fig. 4e), and a strong increase in the Al concentrations towards the end of the monitoring period (Fig. 4f). Bergholm *et al*. (2003) reported a decrease in pH of 0.3–0.3 units and an increase in the concentration of a range of elements in soil water at 50 cm depth in a similar experiment in Sweden. In their experiment, however, the total amount of N and S applied over a 10-year period was 1100 and 1254 kg ha^{-1}, respectively, which is more than 3-fold the levels used in our experiment.

The regular addition of relatively large amounts of sulphate is clearly the main reason for the loss of Ca^{2+}, Mg^{2+}, K^+, and even Al^{3+}, from the organic and uppermost mineral soil layers and their leaching down to a depth of 20 cm: there were highly significant correlations between the concentrations of sulphate and these cations (Table 6). The transport of Al down to a depth of 20 cm is supported by the increase (although not statistically significant) in the exchangeable Al concentration at a depth of 5–10 cm in the mineral soil (Table 4). Bergholm *et al*. (2003) also reported a strong relationship between the Al and sulphate concentration in soil water at a depth of 50 cm. The slight increase (although non significant) in nitrate concentrations in percolation water is interesting because this is widely considered to be a sign of nitrogen saturation of the uppermost soil layers (Gundersen *et al*. 1998). However, the nitrate concentrations in both the C and N + S treatments were extremely small, and in most cases below the limit of quantitation of the analytical instruments. The C/N ratio in the organic and 0–5 cm soil layers were still relatively high (well above 25), and the tree stand continued to show a strong growth response to the annual application of nitrogen up until the end of the experiment, suggesting that N concentrations on the site had not reached the so-called nitrogen saturation level. Bergholm *et al*. (2003) reported appreciable NO_3-N concentrations in soil water at a depth of 50 cm towards the end of a 10-year experiment in a Norway spruce stand involving the application of ammonium sulphate at a level approximately 3-fold higher than that in our experiment. However, the C/N ratio of the organic and uppermost mineral soil layers in the spruce stand in their experiment was already low (between 26 and 20) before the onset of the treatment involving the addition of relatively large amounts of ammonium, and therefore the onset of nitrification is to be expected (Gundersen *et al*. 1998).

Conclusions

The amounts of nitrogen added in the experiment have not led to so-called nitrogen saturation, and

Table 6. Correlation matrix showing the relationship (r^2) between cations and anions in percolation water collected at a depth of 20 cm on the plots treated with annual additions of the N + S (ammonium sulphate) treatment. For the control (C) plots $n = 120$, and for the N + S plots. $n = 154$. *** = $p < 0.001$.

	pH	Al	Ca	Mg	K	NO_3-N
Control						
Al	–0.615***					
Ca	0.583***	–0.235				
Mg	0.100	0.019	0.342			
K	0.295	–0.110	0.488***	0.234		
NO_3-N	0.229	–0.009	0.077	0.096	0.044	
SO_4-S	–0.143	0.402***	0.326	0.178	0.466***	–0.074
N + S						
Al	–0.474***					
Ca	0.278	0.296				
Mg	0.138	0.362	0.852***			
K	0.209	0.364	0.813***	0.766***		
NO_3-N	0.269	–0.082	0.111	0.103	0.219	
SO_4-S	–0.229	0.855***	0.658***	0.691***	0.729***	–0.006

there were only very slight indications of the onset of nitrification. The large leaching losses of Ca and Mg from the surface soil, and subsequent acidification of the uppermost mineral soil layers, were primarily due to the addition of large amounts of sulphate. The growth increase of the stand has also undoubtedly contributed to the decrease in exchangeable Ca and Mg from the organic layer. According to the results for the control plots, it is also clear that the levels of acidifying nitrogen and sulphur deposition in southern Finland during the 1970s and 1980s have not been high enough to cause a threat to site fertility through soil acidification. The treatment with annual applications of nitrogen and sulphur considerably increased stand growth and the litterfall production of Scots pine. The results clearly showed that the pines growing on this very infertile site were, and still are, suffering from a shortage of nitrogen. Fertilization on such sites is not normally recommended owing to the low wood yield and the potential problem of the fertilizers leaching into the groundwater. However, the results obtained in this study are interesting in the light of these recommendations because the growth reaction was reasonable and there was no leaching of nitrate from the uppermost mineral soil layers.

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